EFFECT OF THE FREQUENCY OF WEAK OSCILLATING MAGNETIC FIELDS ON SUPERCOOLING AND FREEZING KINETICS OF PURE WATER AND 0.9% NaCl SOLUTIONS

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ABSTRACT

Several studies in the literature suggest that the frequency of weak oscillating magnetic fields (OMFs) significantly affects supercooling and freezing kinetics, although the effects described by different authors are apparently contradictory. To shed light on this matter, we froze pure water samples and 0.9% NaCl solutions in a 0.8-mT magnetic field at different frequencies (20, 50, 200, and 2000 Hz). Moreover, experiments with no OMF application were also performed to act as controls. Our results show that the application of a 0.8-mT OMF at frequencies between 20 Hz and 2000 Hz during freezing has no effect on the parameters that characterize either supercooling (time at which nucleation occurred and extent of supercooling achieved at the sample center) or freezing kinetics (precooling, phase transition, and tempering times), both in pure water and 0.9% NaCl solutions. More research is needed to elucidate all the factors that could affect OMF-assisted freezing.

Keywords: electromagnetic freezing; oscillating magnetic fields; frequency; supercooling; ice nucleation; freezing kinetics

1. INTRODUCTION

OMF-assisted freezing is an innovative technology that basically involves applying a weak oscillating magnetic field (OMF), usually of low to extremely low frequency, during the freezing process. This technology has already been commercialized and OMF-assisted freezers consist of an OMF generator attached to a conventional quick-freezing unit. Even though OMF-assisted freezers can nowadays be acquired in many countries in the world, the scientific community has not yet found evidence of their efficacy in improving either freezing kinetics or the quality of frozen foods (James, Purnell, & James, 2015a; Otero, Rodríguez, Pérez-Mateos, & Sanz, 2016). Despite

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this, OMF-assisted freezers are publicized as the next-generation blast freezers and a Japanese company (ABI Co. Ltd., Chiba, Japan) commercializes them under the suggesting name of CAS (Cells Alive System) freezers. According to the patents, the magnetic fields applied in these freezers induce forces of magnetic vibration in the water molecules of the product that enhance water supercooling and heat transfer. Supercooling is a phenomenon especially important in the freezing process because it is directly related to the ice nucleation rate. Thus, the larger the extent of supercooling, the larger the amount of ice instantaneously formed at nucleation, the smaller the size of the ice crystals, and the shorter the phase transition time (Zaritzky, 2011). Therefore, in OMF-assisted freezing, small ice crystals are hypothetically formed throughout the product. These small crystals cause limited damage and, thus, the quality of the fresh product remains theoretically unaltered (Owada, 2007; Owada & Kurita, 2001; Owada & Saito, 2010; Sato & Fujita, 2008).

Commercial OMF-assisted freezers usually apply not only oscillating magnetic fields but also static ones. Static magnetic fields (SMFs) are produced by permanent magnets embedded in the walls, ceiling, and floor of the freezing cabinet, while oscillating magnetic fields (OMFs) are generated by magnetic coils arranged around the freezing trays inside the cabinet. The strength values of the magnetic fields applied in commercial freezers are not usually provided by the manufacturers, but some researchers have recently measured them in some CAS freezers (ABI Co., Ltd., Chiba, Japan) for different 'CAS energy' conditions (James, Reitz, & James, 2015b; Otero, Pérez-Mateos, Rodríguez, & Sanz, 2017; Purnell, James, & James, 2017). These measurements revealed that significant spatial magnetic gradients are established throughout the freezing cabinet of CAS freezers that could put into question the uniformity of the pursued effects (Otero et al., 2017). Moreover, the strength of the applied magnetic fields is rather low, only 1-2 orders of magnitude larger than the Earth's natural magnetic field (0.025-0.06 mT). Thus, at the center of the cabinet, Otero et al. (2017) measured strength values of 0.14 mT and 1.4-1.5 mT for the SMF and OMF, respectively, while James et al. (2015b) and Purnell et al. (2017) reported OMF strength values between 0.1 mT and 0.4 mT for different 'CAS energy' conditions. These extremely low strength values cast doubt on the effect that commercial freezers can have on a molecule with low magnetic susceptibility such as water and, therefore, scientific studies on the effects of both static and oscillating magnetic fields on freezing are urgently needed.

To assess the effect of SMFs on supercooling and freezing kinetics, we recently froze 10-mL pure water samples and 0.9% NaCl solutions between two magnets (Otero, Rodríguez, & Sanz, 2018). Even though the distance between the magnets was shorter than 40 mm, significant spatial magnetic gradients were established and, thus, the SMF strength achieved throughout the sample ranged between 107 mT and 359 mT when unlike magnet poles faced each other and between 0 mT and 241 mT when like magnet poles were next to each other. Despite these SMF strengths were considerably larger than those achieved in commercial freezers, no effect on either supercooling or freezing kinetics was detected.

The effect of OMFs on freezing has also been evaluated in the literature. Thus, Watanabe, Kanesaka, Masuda, and Suzuki (2011) reported that 0.5 to 10-mT magnetic fields at 50 Hz have no effect on supercooling of pure water and NaCl solutions. Similar conclusions were drawn by Naito et al. (2012) after freezing distilled and saline water in a 0.5-mT magnetic field at 30 Hz. In real foods, James et al. (2015b) did not detect any effect of OMFs (0.1-0.4 mT, 0-50 Hz) on the extent of supercooling reached by garlic bulbs before freezing, while Suzuki et al. (2009), Purnell et al. (2017), and Rodríguez, James, and James (2017) reported no effect of OMFs on freezing kinetics of different products, including fruits and vegetables (radish, potato, and apple), fish (yellow tail and tuna) and meat (pork loin). However, some authors claim that low-strength OMFs can affect a number of physical properties of water when applied at optimal frequencies. For example, Semikhina and Kiselev (1988) subjected bidistilled water to weak OMFs (up to 0.9 mT and at frequencies between 0.01 and 200 Hz) for 5 h and reported that, for a given OMF intensity, there was an optimal frequency that significantly affected water properties (electrical conductivity, speed of sound or refractive index, among others) and produced maximal supercooling. In this sense, Mihara et al. (2012) and Niino et al. (2012) also reported an effect of the OMF frequency on supercooling. They froze physiological saline solutions in a 0.12-mT OMF at frequencies from 50 Hz up to 200 kHz and observed that the extent of supercooling before nucleation could be significantly increased by using frequencies larger than 200 Hz. By contrast, Zhan, Zhu, and Sun (2019) froze physiological saline samples (9 g/L NaCl solution) and found that a 10-mT OMF at 200 Hz significantly reduced supercooling before nucleation but, at 100 Hz, the extent of supercooling increased when poly dimethyl diallyl ammonium chloride was added (10 mmol/L solutions with physiological solution as the base).

Recently, our research group has designed, built, and characterized an electronic device able to generate OMFs of, at least, the same strength as those applied in commercial CAS freezers, but of a considerably wider range of frequencies. Experimental measurements and modeled results showed that the OMF strengths generated inside the coil of this device were very homogeneous and, therefore, it could be used to evaluate the effects of OMFs on freezing with confidence (Rodríguez, 2017). Therefore, this electronic device has been employed in this paper to evaluate the effect of the frequency of weak OMFs on freezing. To do so, two types of freezing experiments were designed to focus on the effects of OMFs either on supercooling and ice nucleation or on freezing kinetics. In both types of experiments, pure water samples and 0.9% NaCl solutions were frozen in a 0.8-mT magnetic field at different frequencies (20, 50, 200, and 2000 Hz). These specific OMF characteristics were chosen because they cover the range employed in commercial freezers (0.1-1.5 mT and up to 50 Hz) and also that at which Mihara et al. (2012), Niino et al. (2012), and Zhan et al. (2019) reported effects on supercooling (0.12-10 mT and 200 Hz and higher). Moreover, experiments with no OMF application were also performed to act as controls.

The results obtained in this paper contribute to shedding light on the effects that the frequency of weak OMFs have on supercooling and freezing kinetics and provide new information for discussing the effectivity of OMFs in improving food freezing.

2. MATERIALS AND METHODS

2.1. Samples

Ultrapure water (type I, Milli-Q system, Millipore, Billerica, MA, USA) and 0.9% NaCl (Panreac Química SA, Barcelona, Spain) solutions were used in this study. All the samples were located in glass vials closed with polypropylene caps. The sample and vial volume depended on the type of freezing experiment. As small samples are more easily supercooled than large ones (Zaritzky, 2011), small samples of 1.5 mL, located in 2-mL vials, were employed in the supercooling experiments. However, in these small samples, freezing times are very short and, therefore, small experimental errors can produce large variability in the results. For this reason, in the freezing kinetics experiments, larger samples of 6 mL, located in 12-mL vials, were used.

Before each freezing experiment, the samples were tempered in a circulating water bath (model TC-102, Brookfield, Ametek Inc., Middleboro, MA, USA) for, at least, 30 min to achieve a uniform initial temperature of 25 ± 0.5 °C. After each freezing experiment, the frozen samples were discarded (the samples were not reused).

2.2. Freezing experiments

Freezing experiments were performed in a 100-liter chest freezer (AFG 050 MAP, Whirlpool Corp., Benton Harbor, MI, USA) set at −27 ± 1 °C. Inside this freezer, a lab-built OMF generator, thoroughly described by Rodríguez (2017), was installed. In brief, the OMF generator consisted of an air core inductor for generating OMFs in a wide range of frequencies and a frequency-variable single-phase inverter device for converting the voltage supplied by a DC power source into a sinusoidal current. The air core inductor was a Litz-wire solenoid of 75 turns distributed in one layer around a bobbin (5.75 cm radius and 8.6 cm high). The inverter was able to work in a wide range of frequencies thanks to adequate strategies for the driving of switches at high and low frequencies programmed in a commercial microcontroller unit. The setup was completed by a fan (5958, ebm-papst Inc, Mulfingen, Germany), installed behind the air core inductor, to force air circulation inside the freezer.

In each freezing experiment, one sample was placed at the central position of the solenoid to be frozen either with or without OMF application. In OMF experiments, the RMS current through the coil was adjusted to 1.47 A to induce an OMF RMS strength of 0.8 mT. At these conditions, the active power was 2.14 W (equivalent series resistance of the coil = 990 mΩ). Moreover, OMF frequencies were set at 20 Hz, 50 Hz, 200 Hz, and 2000 Hz in OMF_20Hz, OMF_50Hz, OMF_200Hz, and OMF_2000Hz experiments, respectively. In control experiments (C experiments), no current was circulated through the coil and, therefore, no OMFs were applied. To simulate the heat dissipated in OMF experiments as a consequence of the Joule effect, 3 electric heating wires in parallel, with a total resistance of 56 Ω, were put at 10.9 V DC to get the same active power and temperature conditions in the freezer as those of OMF experiments.

As previously mentioned, two types of freezing experiments were performed to study the effect of the OMF application on either supercooling or freezing kinetics. During the supercooling experiments, 2 fiber-optic sensors (T1S, Neoptix Inc., QC, Canada), connected to a signal conditioner (Reflex-4 Neoptix Inc., QC, Canada), were employed to measure the temperature at the geometric center of the sample and inside the freezer. For each condition tested, the temperature was recorded at the sample center to detect the extent of supercooling achieved before nucleation, while the temperature inside the freezer was recorded to verify that all the freezing experiments were performed at the same temperature. All the temperature measurements were monitored and recorded every second via the manufacturer software (Optilink-II, Neoptix Inc., QC, Canada). Supercooling experiments were considered finished 1 minute after the nucleation occurred and they were independently repeated 25 times for each condition.

In the freezing kinetics experiments, the sample volume was larger and, therefore, an additional fiber-optic sensor (T1S, Neoptix Inc., QC, Canada), located at the sample surface, was needed to easily detect the time at which nucleation took place. In these experiments, all the temperature measurements were monitored and recorded every 5 seconds. Freezing experiments were considered finished when the sample center reached −20 °C and they were independently replicated, for each condition, 15 times.

2.3. Analysis of the freezing experiments

After the freezing experiments, all the temperature curves were analyzed and their first derivatives were obtained by using the software Matlab (v. 7.11.0.584 (R2010b), MathWorks Inc., Natick, MA, USA). Figure 1 depicts an example of a typical freezing curve, its first derivative, and all the parameters analyzed in this paper.

Freezing experiments in 1.5-mL samples were examined to assess the effect of the OMF application on the extent of supercooling, ΔT (°C), attained before nucleation. The time at which nucleation occurred, t_{nuc} (s), was identified by a sudden drop in the slope of the freezing curve due to the release of latent heat (Rahman et al., 2002). In this paper, T_c^{nuc} (°C) was defined as the minimum temperature at the sample center just before nucleation. When T_c^{nuc} was lower than the freezing point of the sample (T_{fp}: 0 °C and −0.6 °C for pure water and 0.9% NaCl solutions, respectively), the extent of supercooling attained at the sample center was $\Delta T_c = T_{fp} - T_c^{nuc}$. In other cases, no supercooling existed at the sample center and ΔT_c was zero.

Freezing experiments in 6-mL samples were analyzed to assess the effect of the OMF application on freezing kinetics. To do so, the duration of the 3 key steps of the freezing process (precooling, phase transition, and tempering) was evaluated. The precooling time, t_{prec} (s), was defined as the time span between the onset of the experiment and nucleation and, therefore, $t_{prec} = t_{nuc}$. The phase transition time, t_{pt} (s), was calculated as the time span between nucleation and the end point of freezing. The end point of freezing was identified as the maximum observed, after the freezing plateau, in the slope of the freezing curve recorded at the sample center (Rahman et al., 2002). The tempering time, t_{temp} (s), was the time span between the end point of freezing and the moment at which the sample center reached −20 °C.

2.4. Statistical analysis

The statistical analysis of the characteristic parameters recorded to assess the effect of the OMF frequency on supercooling (t_{nuc} and ΔT_c) and freezing kinetics (t_{prec}, t_{pt}, and t_{temp}) was performed using the software program IBM SPSS Statistics v. 24.0.0.1 for Windows (IBM Corp., Armonk, NY, USA). After checking the prerequisites of normality and homogeneity of variances, a one-way analysis of variance (ANOVA) was carried out to detect whether freezing conditions applied in control and OMF experiments produced significant differences on either supercooling or kinetics of the freezing process. The significance level was set at 5%.

3. RESULTS AND DISCUSSION

3.1. Effect of the OMF frequency on supercooling and ice nucleation

The temperature evolution at the center of 1.5-mL pure water samples during typical C and OMF_200Hz supercooling experiments is depicted in Figure 2. In supercooling experiments, attention is focused on supercooling and ice nucleation and, therefore, the plots in Figure 2 only show the initial steps of the freezing process, that is, the precooling step and part of the freezing plateau. The plots recorded in 0.9% NaCl solutions were similar (data not shown), except for the temperature at the freezing plateau (T_{fp} = −0.6 °C).

Figures 2a and 2b exhibit the typical curves found in C experiments. Even though the experimental conditions were identical in all C experiments, two types of freezing curves were observed due to the stochastic nature of nucleation and the thermal gradients established during the freezing process. Thus, some C experiments showed that, after reaching the freezing point, the temperature at the sample center remained stable on the freezing plateau and; therefore, ΔT_c = 0 (Figure 2a). By contrast, in other C experiments, the temperature at the sample center decreased well below T_{fp} (Figure 2b) and, therefore, large supercooling was observed before the freezing plateau. To understand these results, it is important to note that ice nucleation is a stochastic process that requires a certain supercooling to occur. In general, nucleation is triggered at the sample surface because here the temperature is always lower. Obviously, the later the nucleation occurs at the sample surface, the lower T_c^{nuc} and the larger the extent of supercooling reached throughout the sample. For example, in Figure 2a, ice nucleation occurred early, only 204 s after the onset of the experiment. At this moment, the temperature at the sample center was still over T_{fp} (T_c^{nuc} = 1.7 °C) and, therefore, no supercooling and, consequently, no ice nucleation were observed here (after reaching the freezing point, the temperature remained stable on a plateau). In contrast, in Figure 2b, ice nucleation took place much later, after 319 s. At this moment, the temperature at the sample center was −10.7 °C and, therefore, $\Delta T_c > 0$. As the sample center is the hottest point in the sample, this means that the entire sample was supercooled and, therefore, ice nucleation occurred simultaneously throughout the whole volume of the sample and not only at the surface. When no OMFs were applied, we detected $\Delta T_c > 0$ °C in 23 of 25 experiments in pure water and in 14 of 25 experiments in saline solutions.

Figures 2c and 2d reveal that the application of a 0.8-mT OMF at 200 Hz during the freezing process did not affect the shape or the appearance of the freezing curves of pure water and they were similar to those recorded in C experiments. When other frequencies were applied (data not shown), the freezing curves were not visually affected either and $\Delta T_c > 0$ °C was detected in 24, 23, 23, and 25 of 25 samples in OMF_20Hz, OMF_50Hz, OMF_200Hz, and OMF_2000Hz experiments, respectively. In 0.9% NaCl solutions, these proportions were 11/25, 19/25, 25/25, and 13/25 in OMF_20Hz, OMF_50Hz, OMF_200Hz, and OMF_2000Hz experiments, respectively. These proportions must be considered only as quantitative descriptors of the type of curves registered in the freezing experiments and not as a statistical analysis of the results. This analysis requires more precise considerations as described below.

The detailed examination of all the freezing curves clearly revealed the stochastic nature of ice nucleation, as expected. Thus, Figure 3 shows that ice nucleation did not occur at the same time or after reaching a specific temperature in repeated experiments, but in a time-temperature interval. For example, in C experiments, ice nucleation occurred 197-362 s after introducing the pure water samples in the freezer when the temperature at the sample center was between 1.7 °C and −13.1 °C (Figure 3a). Obviously, the longer the nucleation time, the lower T_c^{nuc} and, therefore, the larger the extent of supercooling achieved before nucleation.

Figure 3 reveals that the application of a 0.8-mT OMF during freezing did not affect, whichever its frequency, ice nucleation and, thus, the time-temperature intervals at which nucleation occurred were similar to those recorded in C experiments, both in pure water (Figure 3a) and 0.9% NaCl solutions (Figure 3b). Accordingly, the statistical analysis of the data showed no OMF effect on the nucleation time or on the extent of supercooling reached before nucleation (Table 1). These results corroborate data in the literature that show that low-strength OMFs (< 10 mT), at the frequency of the mains (50-60 Hz) or lower, do not have any effect on supercooling of pure water or saline solutions (Mihara et al., 2012; Naito et al., 2012; Niino et al., 2012; Watanabe et al., 2011). However, our data do not confirm some previous findings that show that higher OMF frequencies (100 Hz-200 kHz) affect ice nucleation, either enhancing (Mihara et al., 2012; Niino et al., 2012) or hampering supercooling (Zhan et al., 2019) in saline solutions. These apparently contradictory results could indicate that some factors, not considered in this paper, can play a significant role on supercooling and ice nucleation. For example, Zhan et al. (2019) degassed the samples and exposed them to OMFs for 20 minutes before freezing. Both factors, degassing and previous exposition to OMFs, could affect the structure and arrangement of water molecules and be responsible for the results described in their paper.

3.2. Effect of the OMF frequency on freezing kinetics

Figure 4 depicts time-temperature plots recorded in 6-mL saline solutions during typical C and OMF_2000 Hz freezing experiments. When other frequencies were applied, the freezing curves were similar and, therefore, they are not shown. Plots obtained in pure water samples were similar too (data not shown), except for the temperature at the freezing plateau (T_{fp} = 0 °C).

In the experiments described in this section, attention is focused on freezing kinetics and, therefore, the plots in Figure 4 show the entire freezing process with its 3 key steps: precooling, phase transition, and tempering. During the freezing process, significant thermal gradients were established along the sample and, thus, the temperature at the sample surface was always lower than that at the sample center.

In the precooling step, sensible heat was removed from the sample and, therefore, its temperature was lowered. After reaching the freezing point at the sample surface, ice nucleation was not triggered immediately in any case, but cooling continued to a temperature well below T_{fp}. However, at a certain degree of supercooling (ΔT_s), ice nucleation suddenly occurred. This event was easily identified by a sudden decrease in the slope of the freezing curve due to the release of latent heat from the sample. As observed in supercooling experiments, two types of plots were recorded: those, similar to Figures 4a and 4c, in which nucleation occurred before T_c reached the freezing point (or, in other words, ΔT_c = 0) and those, similar to Figures 4b and 4d, in which nucleation was triggered when T_c was below the freezing point (ΔT_c > 0). In pure water, we detected ΔT_c > 0, that is, the entire sample was supercooled in 0, 1, 3, 0, and 0 of 15 experiments in C, OMF_20Hz, OMF_50Hz, OMF_200Hz, and OMF_2000Hz samples, respectively, while, in saline solutions, these proportions were 3/15, 3/15, 6/15, 1/15, and 2/15 in C, OMF_20Hz, OMF_50Hz, OMF_200Hz, and OMF_2000Hz experiments, respectively. As expected, these figures were considerably lower than those observed in the supercooling experiments described in section 3.1. Thus, it is well-known that the sample size is a factor that significantly affects supercooling and, in general, small samples are more easily entirely supercooled than large samples (Zaritzky, 2011). In any case, freezing kinetics experiments confirmed that the application of a 0.8-mT OMF, whichever its frequency, did not affect the time at which ice nucleation occurred, both in pure water and in 0.9% NaCl solutions and, consequently, no significant differences were found in the precooling time of the samples frozen at different conditions (Table 2).

During the phase transition step, the temperature at the sample center remained almost constant at the freezing point, whereas the latent heat of crystallization was removed from the sample. In C experiments, the duration of the freezing plateau ranged between 1420 s and 1545 s in replicated pure water samples, while, in 0.9% NaCl solutions, it was between 1390 s and 1480 s. Figure 5 clearly shows that these time intervals were similar to those recorded in OMF experiments and, thus, Table 2 reveals no significant effect of OMF application on the phase transition time, whichever the frequency applied.

Once most of the water was transformed into ice, sensible heat was removed during the tempering step while the sample was cooled down to the freezer temperature. The data in Figure 6 and Table 2 reveal no effect of the applied OMFs on the tempering step.

The results showed in this paper agree with previous data in the literature (Purnell et al., 2017; Rodríguez et al., 2017; Suzuki et al., 2009; Watanabe et al., 2011) that show no effect of weak OMFs (0.1-0.5 mT) on freezing kinetics when applied at the frequency of the mains or lower (< 50-60 Hz). Moreover, they also confirm the results obtained by Zhan et al. (2019) who did not find significant effects of the application of a 10-mT OMF at 100-250 Hz on t_{prec} and t_{pt} when freezing physiological saline solutions.

4. CONCLUSIONS

All the results obtained in this study reveal that the frequency (20-2000 Hz) of weak OMFs (0.8 mT) has no effect on either supercooling or freezing kinetics of both pure water and 0.9% NaCl solutions. The data also show that the application of a 0.8-mT OMF, whichever its frequency, does not improve the conventional freezing process as no significant differences between C and OMF samples were found for any of the analyzed parameters.

Future research should be focused on stronger OMFs that could have detectable effects on freezing. Moreover, the effects of many factors such as the presence of gas in liquid samples, the sample composition, or its exposition to OMFs before freezing should be evaluated to elucidate the reasons of the controversial results found in the literature.

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FIGURE CAPTIONS

Figure 1 Description of the characteristic parameters of the freezing process obtained from (a) a typical freezing curve, (▬): Temperature at the sample center. (---): Temperature at the sample surface and (b) its first derivative at the sample center. T_{fp}: Freezing point of the sample. T_c^{nuc}: Temperature at the sample center when nucleation occurred.

Figure 2 Temperature evolution (°C) at the center of 1.5-mL pure water samples during supercooling experiments. (a-b): Control experiments with no OMF application. (c-d): OMF_200Hz experiments. ΔT_c: Extent of supercooling reached at the sample center just before nucleation. **(❶)**: Precooling step. (❷): Phase transition step.

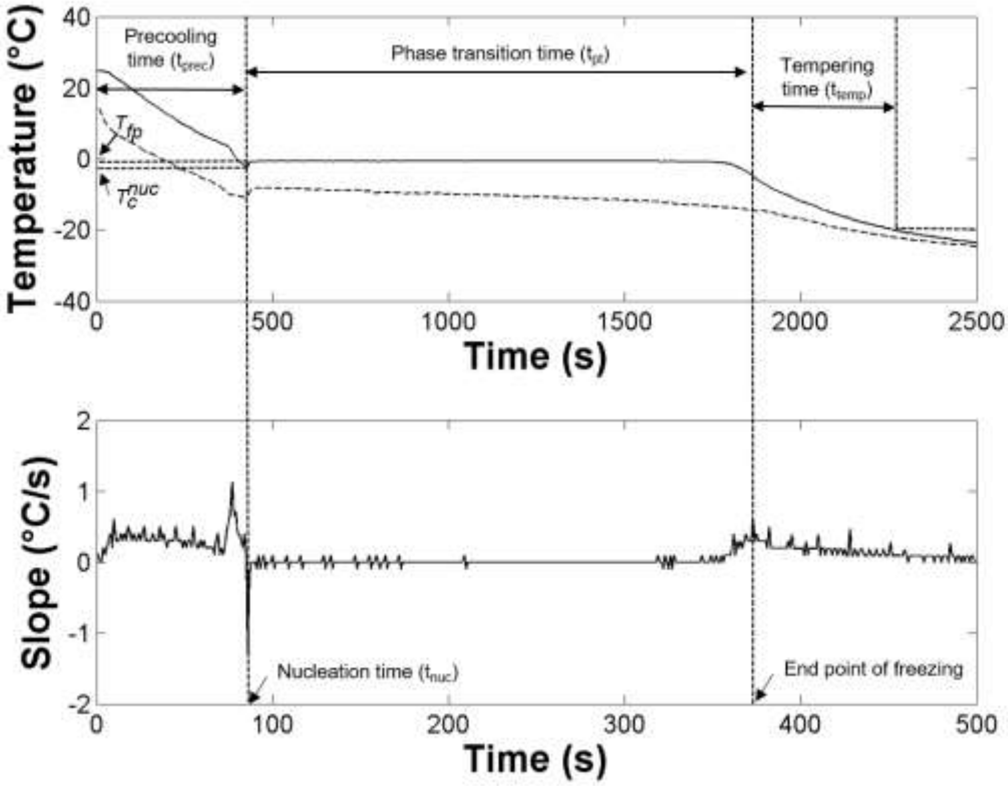
Figure 3 Temperature (°C) and extent of supercooling (°C) at the sample center when nucleation occurred in (+): control, (○): OMF_20Hz, (✳): OMF_50Hz, (□): OMF_200Hz, and (△): OMF_2000Hz experiments. a) Pure water samples and b) 0.9% NaCl solutions.

Figure 4 Temperature evolution (°C) at the surface (---) and center (–) of 6-mL saline solutions during freezing kinetics experiments. (a-b): Control experiments with no OMF application. (c-d): OMF_2000Hz experiments. ΔT_s and ΔT_c: Extent of supercooling reached at the sample surface and center, respectively. **(❶)**: Precooling step, (❷): Phase transition step, and (❸): Tempering step.

Figure 5 Phase transition time (s) in (+): control, (○): OMF_20Hz, (✳): OMF_50Hz, (□): OMF_200Hz, and (△): OMF_2000Hz experiments. a) Pure water samples and b) 0.9% NaCl solutions.

Figure 6 Tempering time (s) in (+): control, (○): OMF_20Hz, (✳): OMF_50Hz, (□): OMF_200Hz, and (△): OMF_2000Hz experiments. a) Pure water samples and b) 0.9% NaCl solutions.

FIGURE 1



Precooling time (t_{prec})
Phase transition time (t_{pt})
Tempering time (t_{temp})
T_{fp}
T_c^{nuc}
Temperature (°C)
Time (s)
Slope (°C/s)
Nucleation time (t_{nuc})
End point of freezing
Time (s)

FIGURE 2

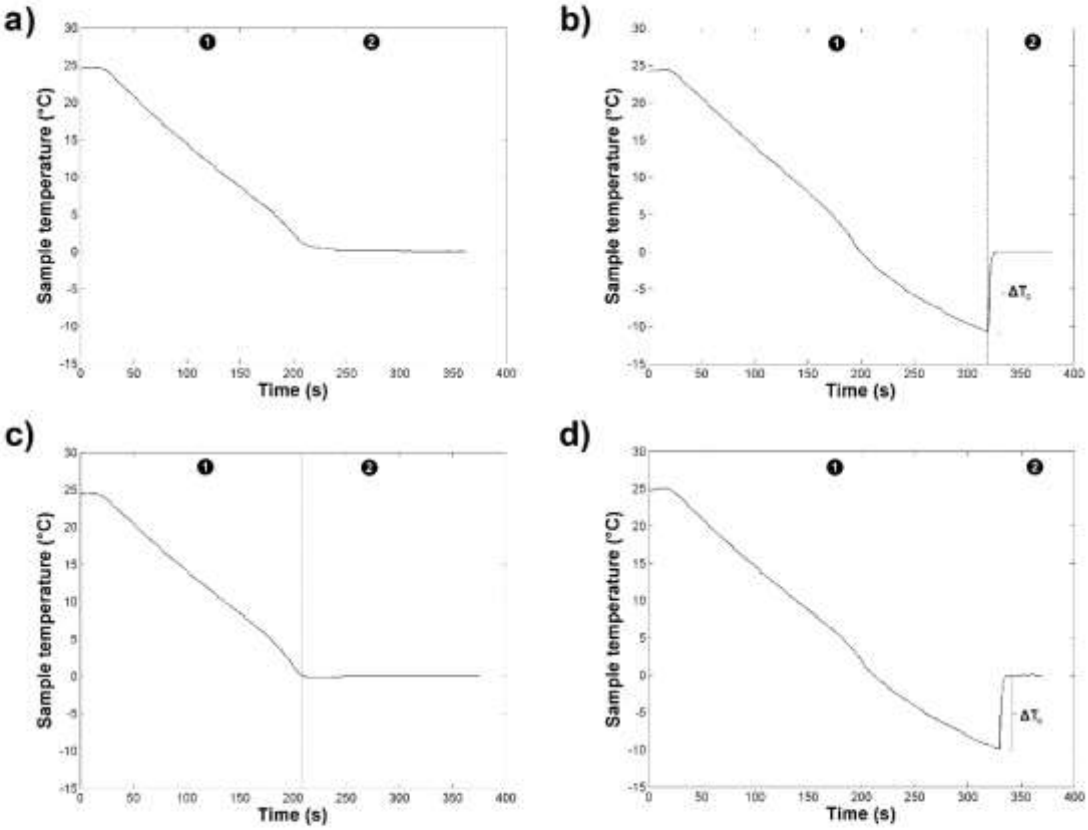
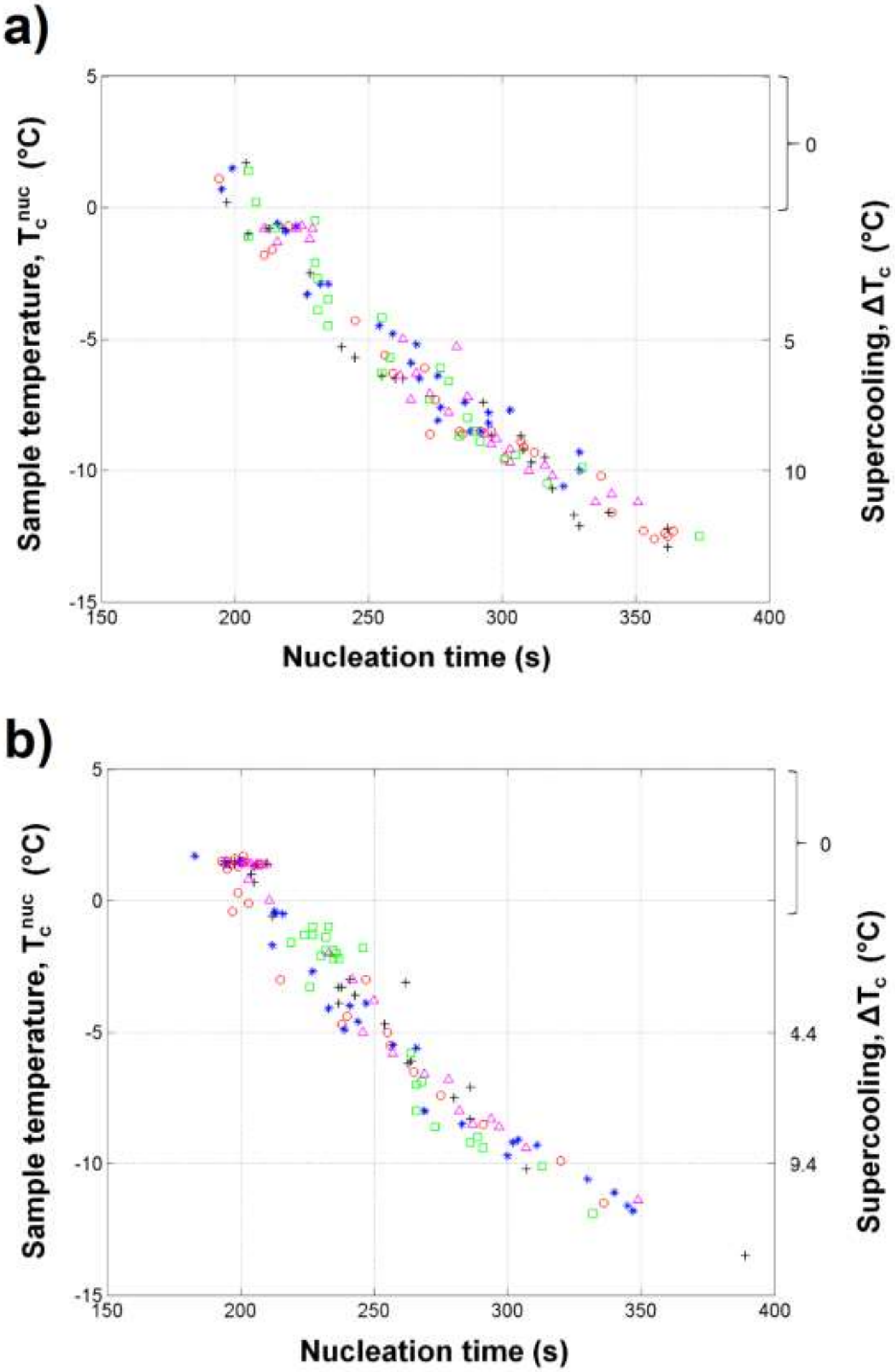


FIGURE 3



a)
Sample temperature, T_c^{nuc} (°C)
Supercooling, ΔT_c (°C)
Nucleation time (s)
5
0
-5
-10
-15
150
200
250
300
350
400
0
5
10
b)
Sample temperature, T_c^{nuc} (°C)
Supercooling, ΔT_c (°C)
Nucleation time (s)
5
0
-5
-10
-15
150
200
250
300
350
400
0
4.4
9.4

FIGURE 4

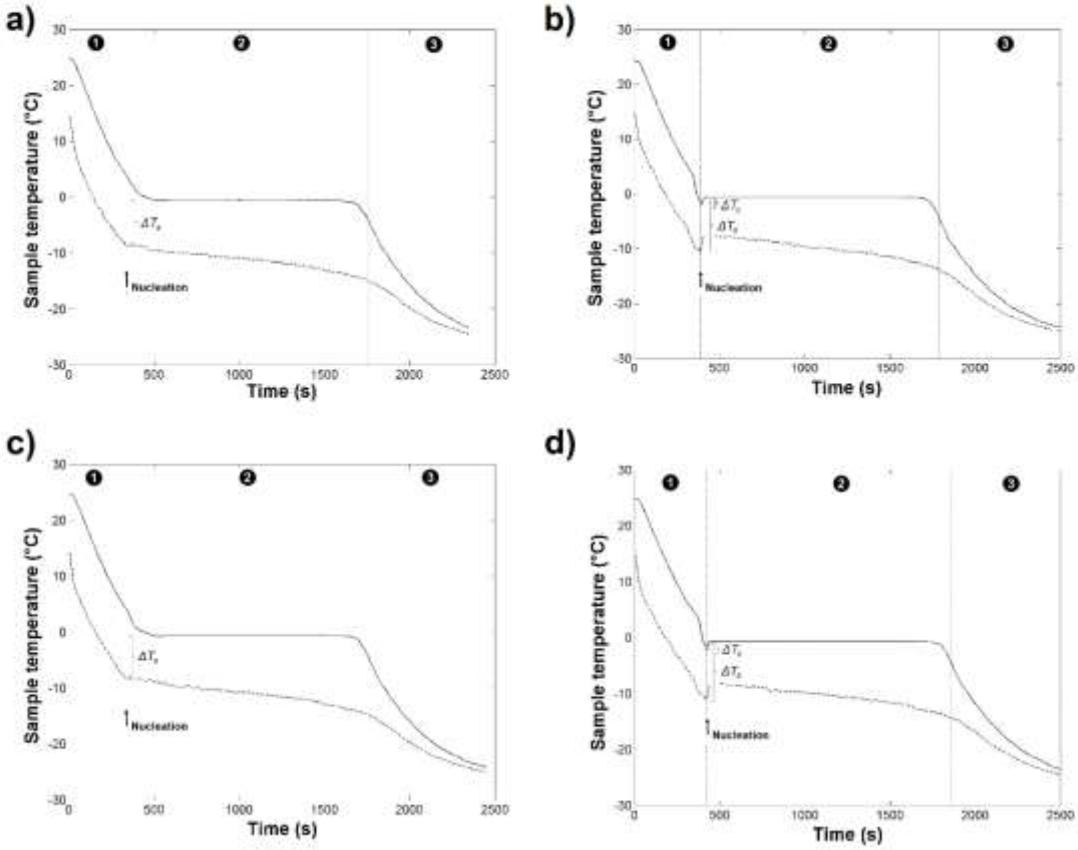


FIGURE 5

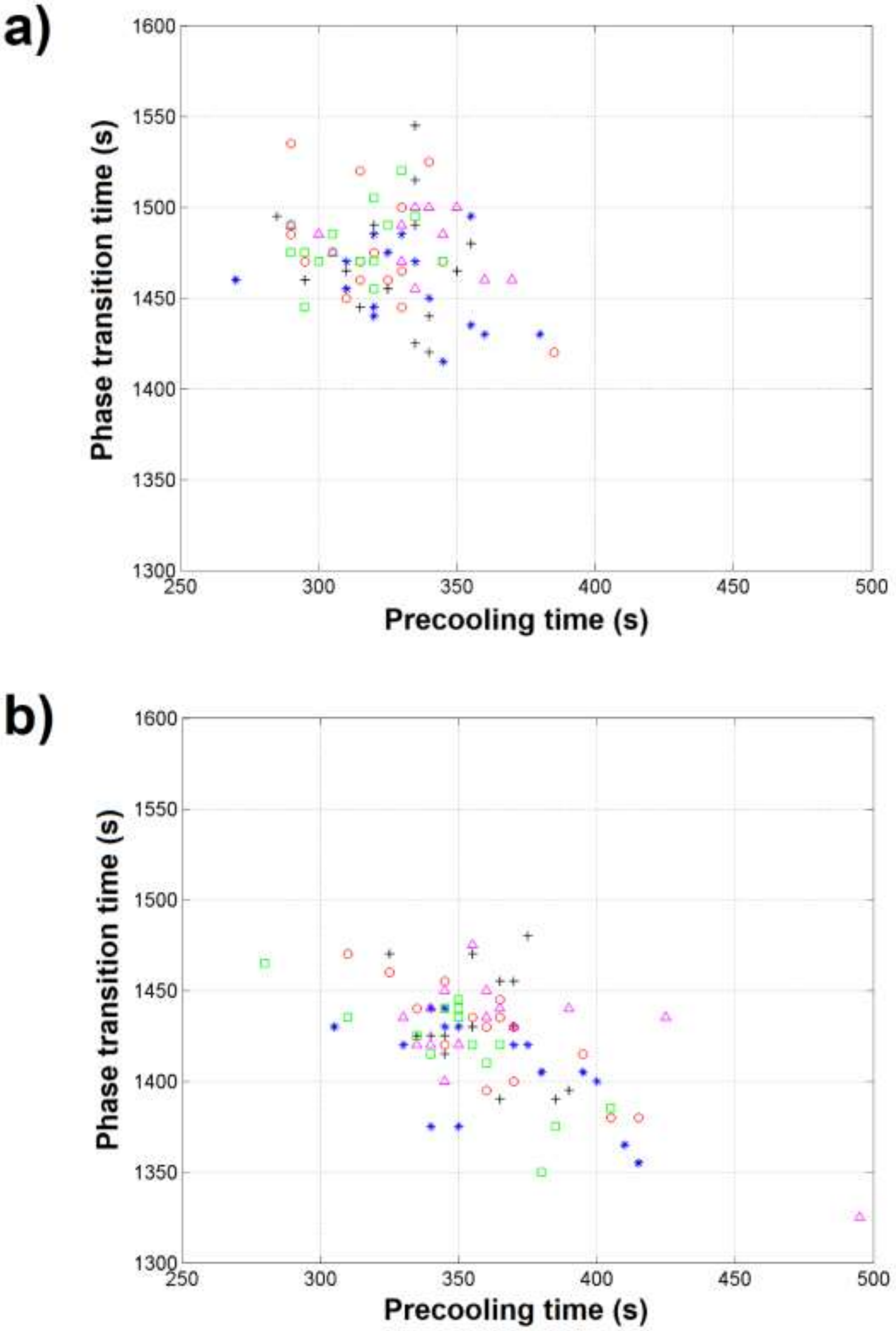

a)
Phase transition time (s)
Precooling time (s)
1600
1550
1500
1450
1400
1350
1300
250
300
350
400
450
500
b)
Phase transition time (s)
Precooling time (s)
1600
1550
1500
1450
1400
1350
1300
250
300
350
400
450
500

FIGURE 6

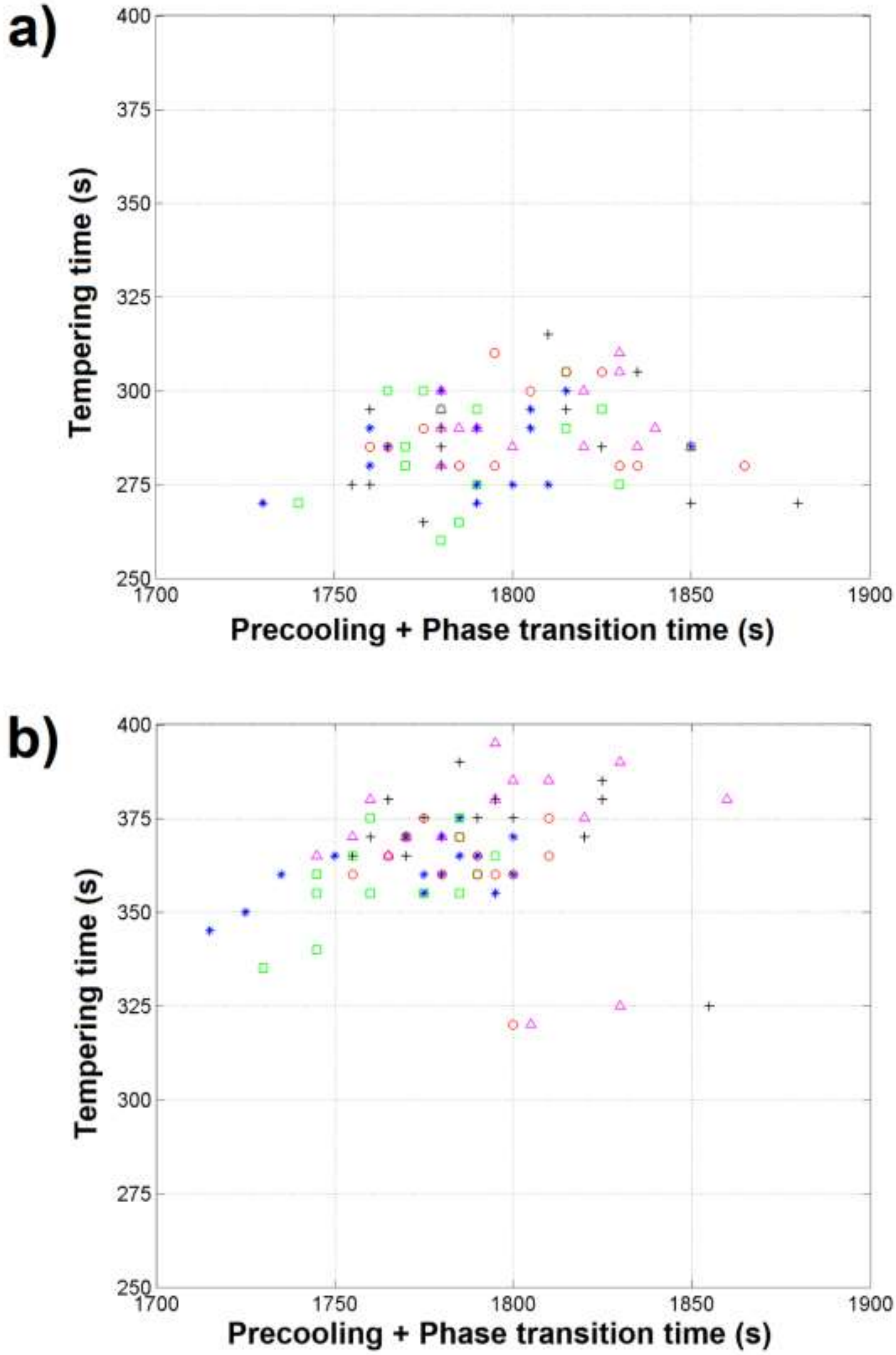

a)
b)
Tempering time (s)
Precooling + Phase transition time (s)
400
375
350
325
300
275
250
1700
1750
1800
1850
1900

TABLE 1 Mean ± standard error values of the main parameters defining supercooling and ice nucleation in freezing experiments with 1.5-mL samples: time (s) at which nucleation occurred (t_{nuc}) and extent of supercooling (°C) reached at the sample center (ΔT_c). For each sample (pure water or 0.9% NaCl solutions), no letters in the same row indicate no significant differences between means due to the OMF applied ($p > 0.05$).

	Pure water					**0.9% NaCl solutions**				
	No OMF	**OMF (0.8 mT)**				**No OMF**	**OMF (0.8 mT)**			
	C	**20 Hz**	**50 Hz**	**200 Hz**	**2000 Hz**	**C**	**20 Hz**	**50 Hz**	**200 Hz**	**2000 Hz**
t_{nuc} (s)	273 ± 11	291 ± 10	265 ± 8	264 ± 9	279 ± 8	240 ± 9	229 ± 8	261 ± 10	253 ± 6	242 ± 9
ΔT_c (°C)	6.5 ± 0.9	7.8 ± 0.8	5.5 ± 0.7	5.7 ± 0.7	6.7 ± 0.7	3.0 ± 0.7	2.5 ± 0.7	5.0 ± 0.8	3.9 ± 0.7	3.2 ± 0.7

TABLE 2 Mean ± standard error values of the main parameters defining freezing kinetics in experiments with 6-mL samples: precooling time (t_{prec}), phase transition time (t_{pt}), and tempering time (t_{temp}). For each sample (pure water or 0.9% NaCl solutions), no letters in the same row indicate no significant differences between means due to the OMF applied ($p > 0.05$).

	Pure water					0.9% NaCl solutions				
	No OMF	OMF (0.8 mT)				No OMF	OMF (0.8 mT)			
	C	20 Hz	50 Hz	200 Hz	2000 Hz	C	20 Hz	50 Hz	200 Hz	2000 Hz
t_{prec} (s)	326 ± 5	322 ± 6	332 ± 7	313 ± 4	326 ± 7	359 ± 5	361 ± 7	363 ± 8	348 ± 8	367 ± 11
t_{pt} (s)	1469 ± 9	1477 ± 8	1456 ± 6	1479 ± 5	1481 ± 4	1432 ± 7	1426 ± 7	1407 ± 7	1420 ± 8	1428 ± 9
t_{temp} (s)	285 ± 4	289 ± 3	284 ± 9	285 ± 3	289 ± 3	372 ± 4	360 ± 4	362 ± 2	360 ± 3	370 ± 6